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The Groningen AMS facility

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Abstract

A new generation accelerator mass spectrometer has been in operation at the Centre for Isotope Research in Groningen, Netherlands since the summer of 1994. It is a 2.5 MV Tandetron, dedicated to radiocarbon (^{14}C) analysis with high precision ($< 0.5\%$). We present here a report for the first year of operation.

1. Introduction

The Centre for Isotope Research of the University of Groningen is a well-established facility for measurement of naturally occurring isotopes. This includes a low-background laboratory for ^{14}C and ^{3}H, and four mass spectrometers for stable isotopes (^{13}C, ^{2}H, ^{18}O, ^{15}N). Since 1994 the laboratory operates a ^{14}C-dedicated accelerator mass spectrometer (AMS), based on a 2.5 MV Tandetron accelerator. The "model 4130" AMS was built by High Voltage Engineering Europe (HVEE) in Amersfoort, Netherlands and is the second of the so-called "new generation" machines [1,2] which is mainly characterised by a simultaneous measurement of all three carbon isotopes. This design allows all three isotopes to be measured under the same conditions.

The layout of the Groningen AMS system is given in Fig. 1. The high intensity Cs sputter ion source produces a C^- beam of typically 50 μA at approx. 36 keV that is focused by an Einzel lens on a defining aperture. After this aperture the beam enters a recombinator [3] that consists of four (second order corrected) 45° magnets. The first pair of magnets analyses the beam and separates the desired masses 12, 13 and 14 at the symmetry plane of the recombinator by about 20 mm. Moveable beam stoppers in the symmetry plane allow every isotope to be injected and analysed separately. A chopper wheel located in the symmetry plane of the recombinator reduces the ^{12}C beam load for the accelerator and the high energy spectrometer by a factor of about 90. The second pair of magnets recombine the three isotopes on the optical axis of the accelerator for simultaneous injection.

In the Tandetron accelerator the singly charged negative ions are focused and accelerated to a terminal, which operates at +2.5 MV. After charge exchange to the mean charge state 3+, the ions are accelerated back to ground potential to an energy of 10 MeV. The high energy acceleration tube has a specially designed, inclined-field electrode arrangement to suppress ambiguities of ions with the same mass–energy product as the wanted 10 MeV $^{14}C^{3+}$ particles, which result from charge exchange processes during the acceleration to ground potential [1].

The final analysis is done by an achromatic high energy spectrometer, consisting of a 110° magnet, a 33° electrostatic deflector and a 90° magnet. The first magnet selects the 3+ charge state and separates the three isotopes. The ^{12}C and ^{13}C beams are measured in individual Faraday cups, located in the image point of the 110° magnet. Typical currents are in the range of 300 to 800 nA. The ^{13}C cup is equipped with an integrated slit system [4]. The error signal that is generated by this system is fed back to the terminal voltage driver and guarantees a stable beam position in the high energy spectrometer at any time.

After the 110° magnet the ^{14}C particles subsequently pass two additional dispersive elements, a 33° electrostatic deflector, which blocks almost all residual ambiguities with the same mass-energy product as 10 MeV $^{14}C^{3+}$, and a 90° magnet for further background reduction, introduced by scattering effects at the 33° deflector plates or by two step charge exchange processes with the residual gas. The latter background is minimised by vacuum conditions below 10^{-8} mbar in the ^{14}C spectrometer. ^{14}C ions are detected in an isobutane filled ionisation chamber kept at a pressure of about 20 mbar isolated from the vacuum system by a mylar foil (≈ 350 $\mu g/cm^2$ thickness). The detector, providing a dE/dx signal and an E signal, is read out by a PC-based multichannel analyser. The ^{14}C peak in the E spectrum is always clearly separated from other mass-14 peaks, as is illustrated in Fig. 2. The ^{14}C count rate is about 100 Hz for a modern sample (100 pMC, or "fraction modern carbon").

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2. Performance

The first results of the Groningen AMS were presented at the 15th International Radiocarbon Conference in Glasgow, UK, in August 1994 [5], almost immediately after the acceptance of the AMS system from the supplier. Performance tests on identical samples (mainly ANU sucrose), and cross checking with conventional ^{14}C decay counting and ^{13}C mass spectrometry showed a precision on $^{13}\delta$ of better than 2‰, and on $^{14}C/^{12}C$ of better than 0.5 pMC, respectively [5].

At present, after just one year of operation, we have measured close to 2000 samples (including many performance test samples, standards and backgrounds). During the year, performance and reliability of the system gradually improved. Recently we started measuring overnight (unattended) runs, greatly enhancing the efficiency of operation.

A typical batch measurement consists of 2 anthracite background targets, 9 ^{14}C standard targets and 24 unknown samples. Each target is measured for 72 "blocks", each block corresponding to 30 s. After every block, data are saved and the $X-Y$ table moves the sample to the next point on the sample surface to be irradiated by the Cs beam, thereby minimising cratering effects. As a working standard for ^{14}C, we have used ANU sucrose, also known as IAEA standard C6 with a ^{14}C content of 150.61 pMC, and a $^{13}\delta_{PDB}$ value of -10.80‰ [6]. For these C6 standards, the measurements for a typical batch result in a precision of 1.5‰ for $^{13}C/^{12}C$, and 3‰ for $^{14}C/^{13}C$. For the time period after acceptance of the machine, i.e. a little more than one year, the variability of the C6 standard is shown in Fig. 3. The ^{14}C results, expressed in fraction modern (pMC) and corrected for isotopic fractionation by means of ^{13}C, are shown as triangles; the statistical errors on these numbers are plotted as circles, and are in the

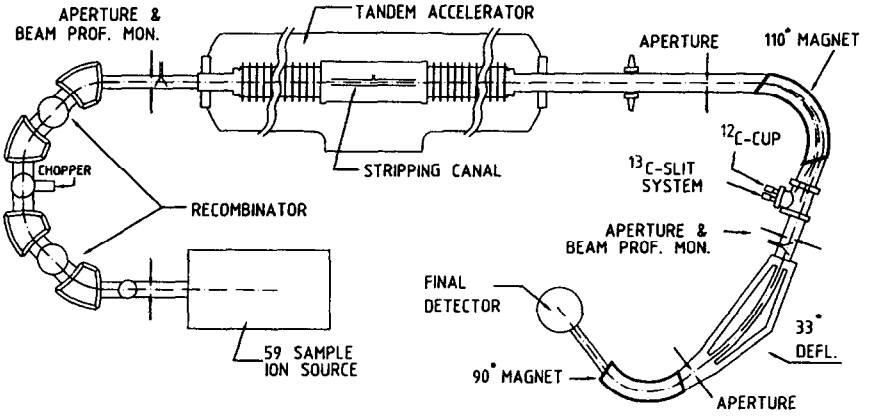

Fig. 1. Schematic lay-out of the Groningen ^{14}C AMS system.

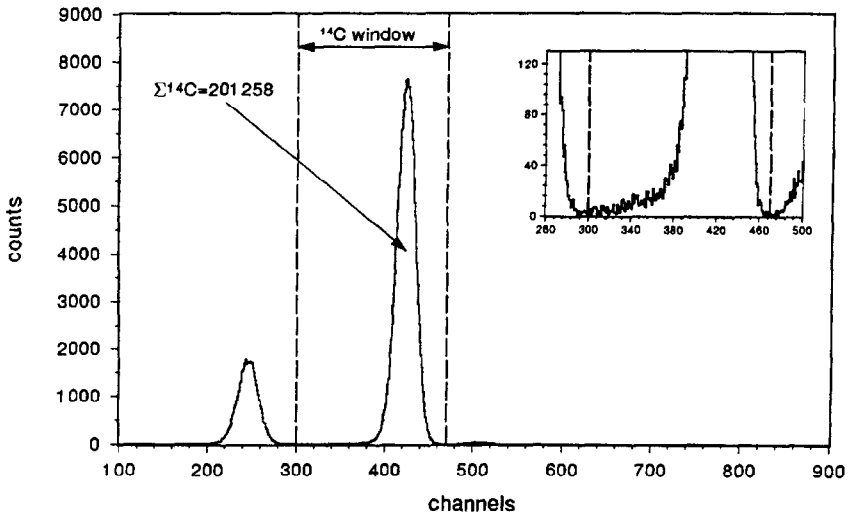

Fig. 2. Ungated ^{14}C energy spectrum from a sucrose sample (150 pMC).

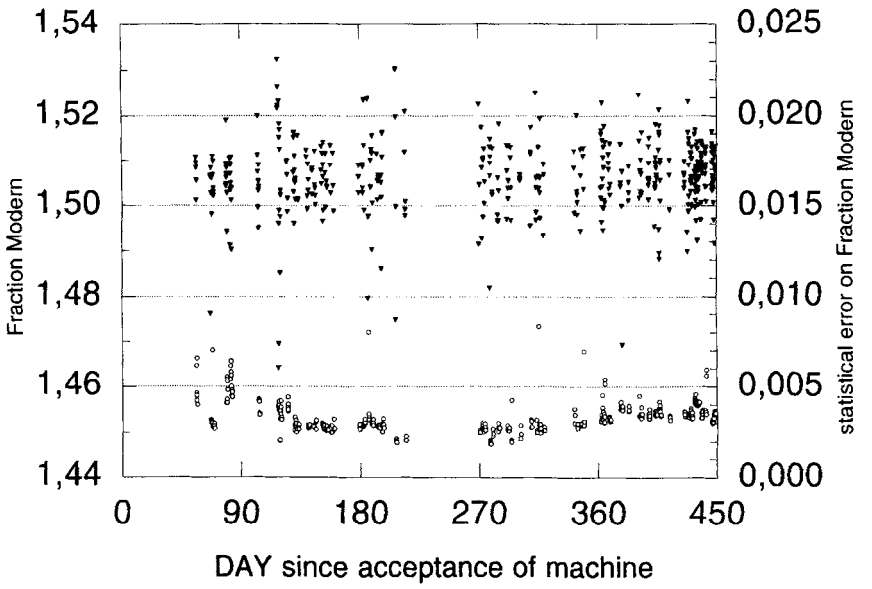

Fig. 3. Sucrose (C6) standard sample measurements during the first year of operation. Triangles: fraction modern (pMC); circles: statistical error.

3–4‰ range. The gap around ordinate 250 corresponds to a period of down time for necessary repairs.

By far the most of the unknown samples measured thus far are not older than 10000 years. We still have to assess a proper background determination. Our present working background material is anthracite of which we assessed an age of 45 ka (this number includes all laboratory treatments). Samples from a supply of natural Ceylon graphite were measured as 52 ka; also natural gas-CO_2 ("Rommenhoeller") which is used in stable isotope mass spectrometry yields a background of around 50 ka. A graphite rod, not handled in the chemistry laboratory and directly pressed into a sample holder, resulted in a background level of 60 ka. The background of the AMS itself (i.e. excluding any target handling) has been checked with a carbon-free aluminium target ("machine blank") and assessed as better than 100 ka [5].

The major source of contamination is at present the combustion. We use an automated elemental CN analyser, consisting of an Cr_2O_3 flash combustion tube, Ag and Cu

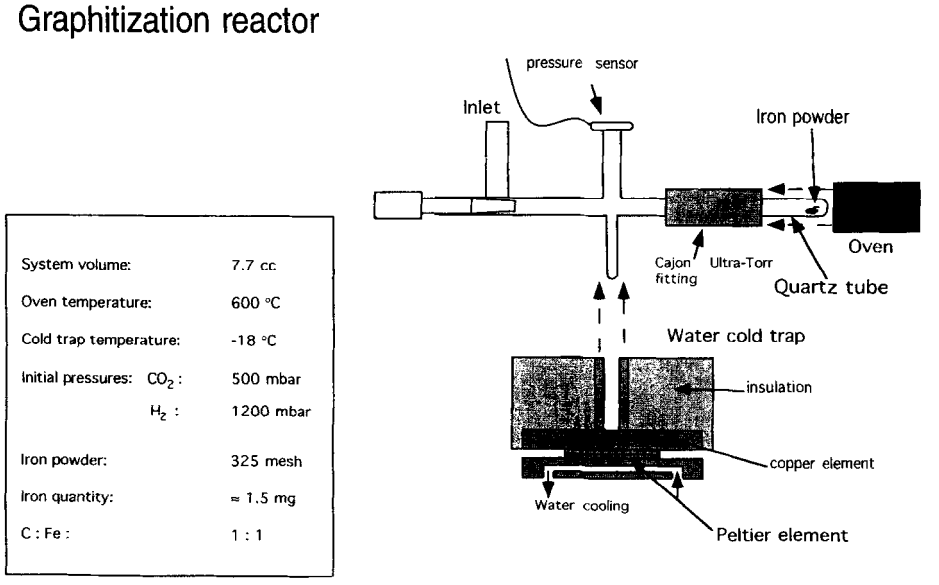

Fig. 4. Graphitisation reactor for AMS samples.

furnaces, a water trap and a gas chromatographic column to separate N_2 from CO_2.

This CO_2 is trapped cryogenically. The CO_2 is converted into graphite by reduction under an excess of hydrogen gas using Fe powder as a catalyst [7,8]. Before reduction, we flush our system with hydrogen in order to remove any CO_2 which might be sticking to the glassware. The Fe powder is pre-reduced with hydrogen. Next, the CO_2 is transferred to a variable volume to adjust the amount of gas. The H_2O is trapped in a cold finger, which is cooled by a Peltier element. The hot side of the element is cooled by water circulation in order to create a temperature of about -18°C. During the reaction, the pressure is monitored on computer screens. In general the reaction takes at least 200 min of time, but usually we graphitise overnight. The graphitisation system contains 10 reactors. One of the reactors is shown in Fig. 4. Finally, the graphite powder is pressed into a target holder, stored in vacuum or argon, and eventually loaded in the ion-source target carousel for measurement.

To test the contamination from this system, we performed a test by alternating graphitising of anthracite and sucrose ("sandwich test") which mimics the worst possible case. The results, for the 5 anthracite samples are shown in Fig. 5, clearly indicating a small "memory effect" due to the sucrose (omitted in the plot) in between.

3. Projects

In the first year of operation, close to 1000 unknown ^{14}C samples were measured. Some projects are extensions of our conventional research program; most are new, employing the new capabilities offered by AMS.

In *hydrology*, a large program is started for isotopic research in Botswana, where for ^{14}C amounts of water only possible with AMS are available. Together with stable isotopes (^{2}H, ^{13}C, ^{18}O) unique information on water-resource development in the region will become available.

In *oceanography*, we participate in some WOCE activities. At present we have measured ^{14}C extracted from ocean water profiles in the Philippine Sea. We have also constructed a special system for ocean-DOC analysis [9]. The first experiments on the North Sea are presently performed.

Atmospheric research has been devoted mainly to a system for "event trapping" at Kollum, a station set-up 30 km northwest of Groningen for off-line isotopic analysis of tropospheric CO_2 (^{13}C and ^{18}O by MS, and ^{14}C by AMS). First results are presented in Ref. [10].

In the *earth* sciences, the main projects under investigation are high-resolution studies of peat deposits. At the Hallstatt plateau (around 500 B.C.) studied at a site in the Netherlands (Egbertsdijkveen) we have detected a reservoir effect and shown that AMS wiggle matching with peat cores is possible [11]. Similar data are obtained at the 9600 B.C. plateau (de Borchert, Netherlands), Lake Gosciaz (Poland) and Lake Suigetsu (Japan). Measurements are in progress, and results are presently being analysed.

In *archaeology*, many dating projects are obviously performed as in any AMS laboratory. Most materials are charcoal and bone. A special project concerns hair, skin, bone and cloth fragments from bodies found in peat bogs around the Dutch/German border area. Amino acids extracted from bones will be measured in Oxford [12]. An archaeological site of utmost importance is ancient Jericho. High precision dates ($\sigma < 20$ years) were obtained by our

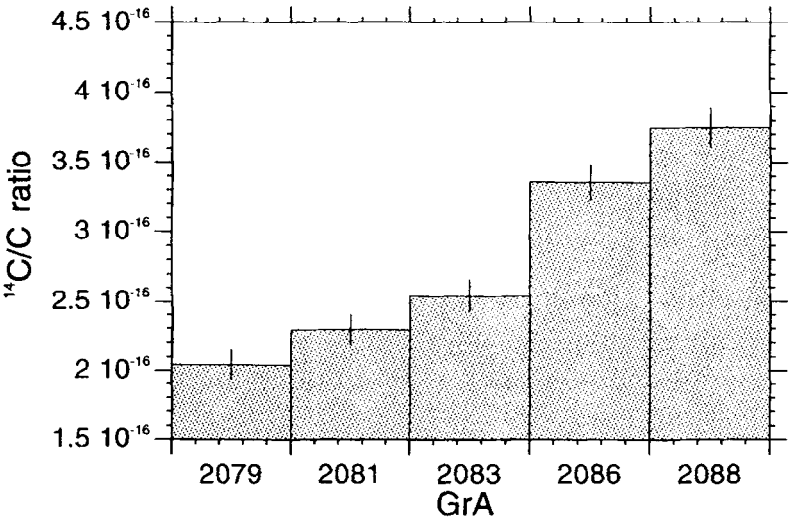

Fig. 5. Results from a memory effect test of the graphite system: $^{14}C/C$ ratio versus sample number for anthracite. In between each anthracite, a sucrose (C6) sample was graphitised.

conventional laboratory. Samples too small for the proportional counters, however, have been measured by AMS [13].

A new application has been developed in *soil* sciences. A new Soil Organic Matter (SOM) fractionation method was developed recently and applied to both maize and pasture. The results indicate that fractions exist with ages ranging from 2 radiocarbon years (coarse fractions) to more than 3000 years (mineral associated with carbon). Maize cropping induced a strong rejuvenation of the finer fractions which represent colloidal organic material in association with soil particles. Because of the small sizes of the fractions, only AMS can be applied; this offers new possibilities for studying SOM dynamics in soils [14].

4. Conclusions and outlook

For a little more than one year, a new generation of Accelerator Mass Spectrometer (AMS) has been in operation at the Centre for Isotope Research in Groningen, Netherlands. The AMS is dedicated to ^{14}C. About 2000 samples (of which 1000 unknowns) have been measured in the first year. The precision in detection ^{14}C is better than 0.5 pMC, and present background is assessed at 45 ka (including all laboratory treatments). The AMS will be heavily used in a variety of applications. In the nearby future, the capabilities of the facility will be extended in the following areas:

1. small AMS samples (the present limit is 1 mg);
2. background reduction – in particular the sample preparation;
3. enrichment to improve counting statistics and possibly extend dating range.

Finally, a database is being installed, containing all information from ^{14}C forms already existing for the conventional laboratory [15], greatly enhanced with all relevant AMS results. The database is truly relational with heavy internal use for administrative and scientific purposes. A future internet browser is considered.

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